Multi-Step TPSR/QMS Technique to Study the Kinetics of NH_3-SCR

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1. What is NO_x?

- Nitric oxides are reactive gases, primarily NO (>90 %) and NO_2.
- They are pollutants involved in many atmospheric processes e.g. formation of photochemical smog and acid rain.
- They are produced as a result of high temperatures during the combustion of fuels.
- Legislation is in place to reduce NO_x emissions i.e. the European Waste Incineration Directive (WID) regulates activities that involve burning or gasification of waste(**Figure 1**).

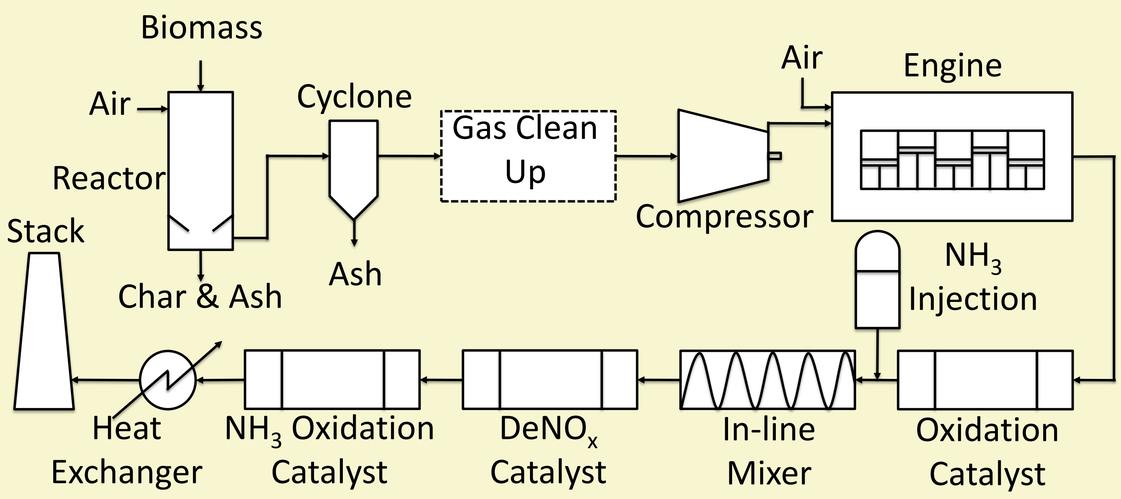

Figure 1. Schematic of proposed biogas engine exhaust treatment system

2. $DeNO_x$ Process

- NH_3-Selective Catalytic Reduction (SCR) is an efficient, established method for NO_x removal. The desired reactions are: $4NH_3 + 4NO + O_2 \rightarrow 6H_2O + 4N_2$ $8NH_3 + 6NO_2 \rightarrow 12H_2O + 7N_2$
- BUT there are some disadvantages including:
  - Ammonia slip
  - Size of the installation
  - Thermal deactivation
- Structured reactors based on metallic short channel structures (**Figure 2**) demonstrate improved mass and heat transfer properties [1] and can remedy these issues [2].

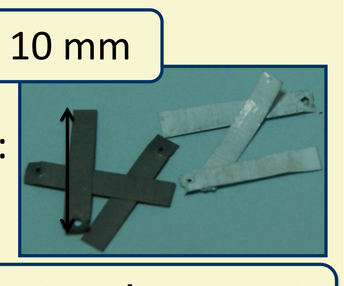

Figure 2. Metal sheet catalyst supports

3. Catlab

- Hiden Analytical CATLAB-PCS microreactor (**Figure 3**) with integrated quadrupole mass spectrometer (QMS) system.
- QMS is a convenient and powerful method for analysing complex reaction mixtures. However, careful calibration is required to obtain reliable, quantitative data.
- There are a number of factors to consider when optimizing the analysis system for a particular application, including: detector selection, ion fragment patterns and relative sensitivity values.

Figure 3. Hiden Analytical CATLAB-PCS microreactor

4. TPSR/TPD Studies

- Coupled with Temperature Programmed Desorption (TPD), Temperature-Programmed Surface Reactions (TPSR) can be used to gain new insights into the mechanism of NO reduction on copper-exchanged zeolites. Results may also shed light on the active centres of the reaction and relative reactivities of surface and gaseous species.
- TPD is used to study the adsorption/desorption of NH_3 and NO (**Figure 4**).
- TPSR can be used to investigate reactions between adsorbed and gaseous species **(Figure 5).**

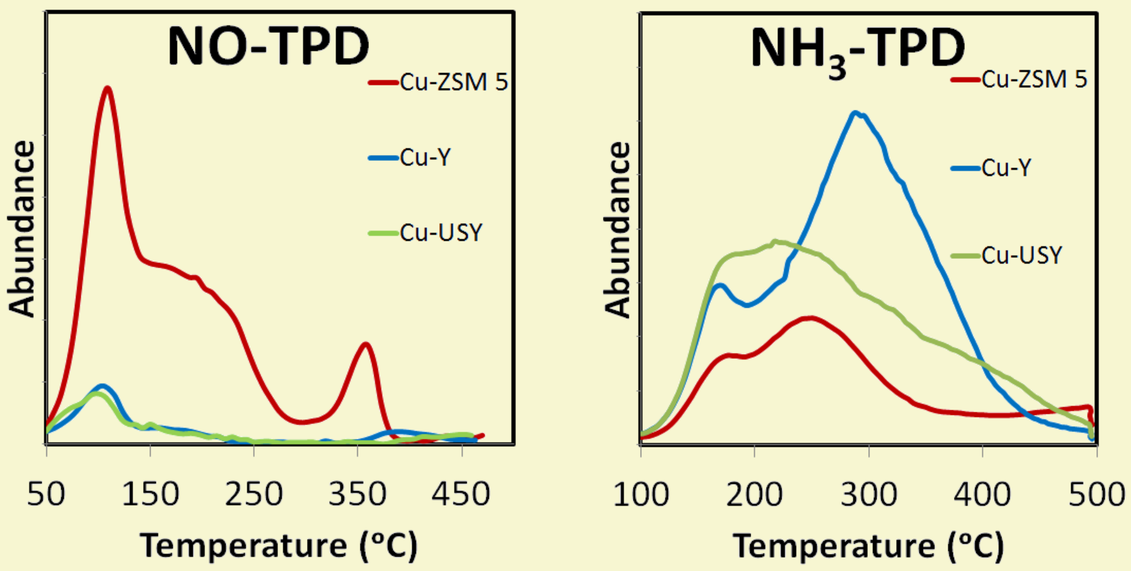

Figure 4. NO-TPD and NH_3-TPD plots of 3 copper exchanged zeolite powders.

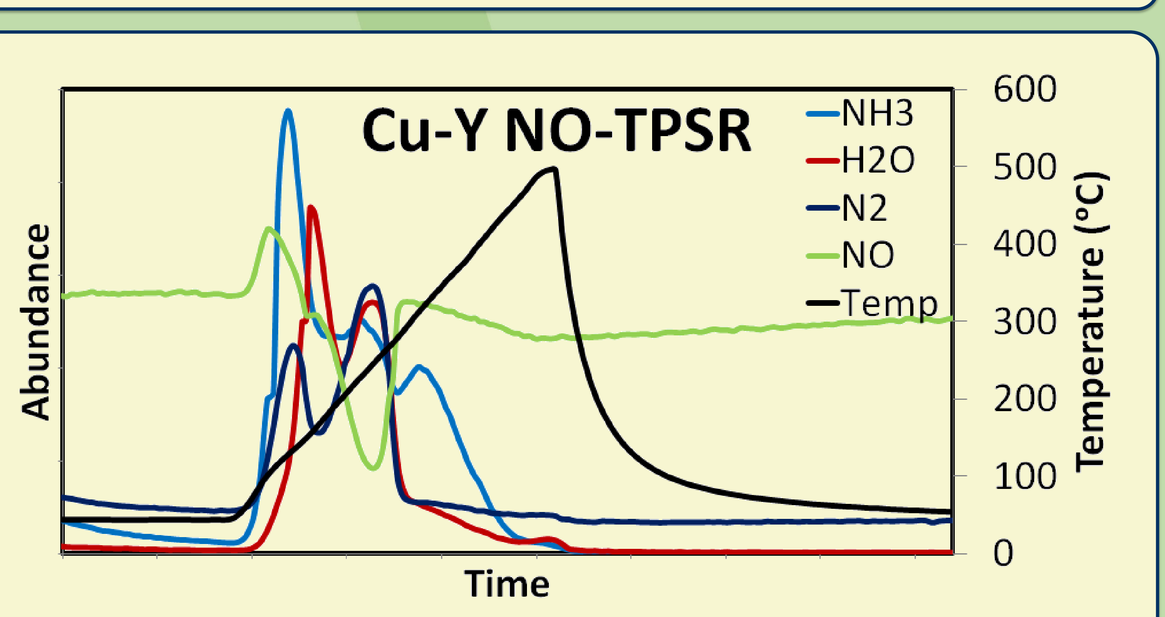

Figure 5. Example NO-TPSR plot obtained from Cu-Y zeolite powder.

6. Conclusions

- Initial results suggest a Eley-Rideal-type mechanism i.e. adsorbed NH_3 reacts with gaseous NO on Cu^+ active centres in the zeolite cavities.
- Obtained data was used for kinetic modelling [3].

REFERENCES
[1]Kołodsiej, A. & Łojewska, J. 2009. *Catalysis Today, 147*, ***S120-S124.***
[2]Ochońska, J. *et al.* 2011 "Copper exchanged zeolite in structured reactor based on wire gauzes for ammonia reduction of nitrogen oxides from biogas turbines", NOEA 2011.
[3]Jodłowski, P. *et al*. 2011 "Modeling of structured reactor based on wire gauzes and zeolite catalyst for ammonia reduction of NOx biogas turbine", NOEA 2011

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